

Spectral absorption of solar radiation by aerosols during ACE-Asia

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[1] As part of the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia), the upward and downward spectral solar radiant fluxes were measured with the Spectral Solar Flux Radiometer (SSFR), and the aerosol optical depth was measured with the Ames Airborne Tracking Sunphotometer (AATS-14) aboard the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft. In this paper, we examine the data obtained for two cases: a moderately thick aerosol layer, 12 April, and a relatively thin aerosol case, 16 April 2001. On both days, the Twin Otter flew vertical profiles in the Korean Strait southeast of Gosan Island. For both days we determine the aerosol spectral absorption of the layer and estimate the spectral aerosol absorption optical depth and single-scattering albedo. The results for 12 April show that the single-scattering albedo increases with wavelength from 0.8 at 400 nm to 0.95 at 900 nm and remains essentially constant from 950 to 1700 nm. On 16 April the amount of aerosol absorption was very low; however, the aerosol single-scattering albedo appears to decrease slightly with wavelength in the visible region. We interpret these results in light of the two absorbing aerosol species observed during the ACE-Asia study: mineral dust and black carbon. The results for 12 April are indicative of a mineral dust-black carbon mixture. The 16 April results are possibly caused by black carbon mixed with nonabsorbing pollution aerosols. For the 12 April case we attempt to estimate the relative contributions of the black carbon particles and the mineral dust particles. We compare our results with other estimates of the aerosol properties from a Sea-Viewing Wide Field-of-View Sensor (SeaWiFS) satellite analysis and aerosol measurements made aboard the Twin Otter, aboard the National Oceanic and Atmospheric Administration *Ronald H. Brown* ship, and at ground sites in Gosan and Japan. The results indicate a relatively complicated aerosol mixture of both industrial pollution (including black carbon) and mineral dust. This underscores the need for careful measurements and analysis to separate out the absorption effects of mineral dust and black carbon in the east Asia region.

INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0360

Atmospheric Composition and Structure: Transmission and scattering of radiation; 3359 Meteorology and Atmospheric Dynamics: Radiative processes; 9320 Information Related to Geographic Region: Asia;

KEYWORDS: light absorption, single-scattering albedo, ACE-Asia aerosols

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1. Introduction

[2] The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) was carried out during March–

May 2001 [Huebert *et al.*, 2003]. Its purpose was to investigate the outflow of aerosols from east Asia during the spring months with particular emphasis on mineral dust outbreaks and air pollution transport. The ACE-Asia intensive field program consisted of measurements made aboard several aircraft (including a Twin Otter and a C-130), aboard the National Oceanic and Atmospheric Administration (NOAA) ship *Ronald H. Brown*, and at ground stations at Gosan, Korea, and various locations in Japan. Measurements were also made in conjunction with several satellite sensors (e.g., Sea-Viewing Wide Field-of-View Sensor (SeaWiFS), Moderate-Resolution Imaging Spectroradiometer (MODIS), Multi-angle Imaging Spectroradiometer (MISR), Advanced Very High Resolution Spectrometer

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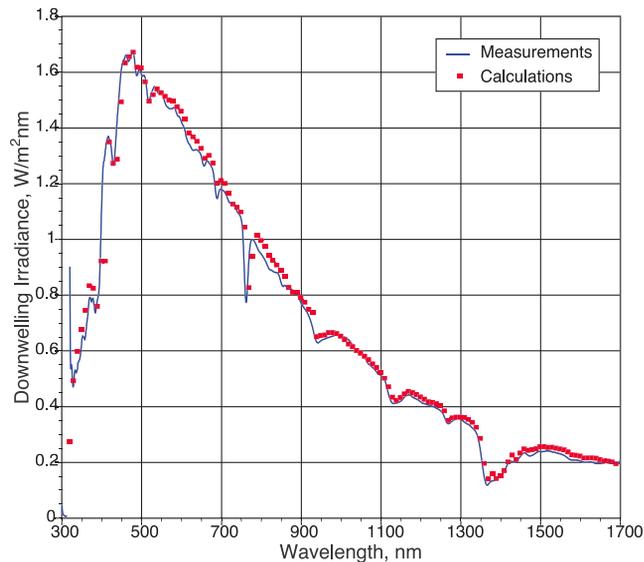


Figure 1. Measured and calculated downward flux at 2 km above the surface of 12 April 2001.

(AVHRR). The study was also coordinated with the Asian Atmospheric Particle Environmental Change Study (APEX program) [Nakajima *et al.*, 2003] and the Transport and Chemical Evolution over the Pacific (TRACE-P) study [Jacob *et al.*, 2003]. For an overview of the ACE-Asia study, see Huebert *et al.* [2003].

[3] The initial results of the ACE-Asia study have been described in a number of papers (*Journal of Geophysical Research*, 108(D23), 2003). During the study period, aerosols from pollution sources, biomass burning and mineral dust from the inland Chinese deserts were transported from the interior to the coast of east Asia. Significantly, several major outbreaks of mineral dust occurred during March and April 2001. One of the most important findings of the ACE-Asia study is that the aerosol particles off the coast of east Asia were complex mixtures of mineral dust, pollution particles, and sea salt. These combinations of particle types are highly variable and make attribution of the aerosol radiative properties to specific aerosol types difficult without detailed measurements.

[4] We report here on the characterization of the solar spectral irradiance fields and the aerosol radiative properties prevalent during two distinct aerosol cases over the Korean Strait during the ACE-Asia study period. The first case on 12 April 2001 was characterized by a moderately thick layer of pollution and dust aerosols while the second case on 16 April had a relatively thin aerosol layer. Net solar irradiance spectra obtained at multiple levels above and below the aerosol layer provided the means to derive the layer absorption spectra. Optical depth measurements were used with the absorption spectra in a radiative transfer model to estimate the spectral absorption optical depth, layer absorption coefficient and single-scattering albedo. These results are interpreted as to the relative contributions of black carbon and dust particles, the two aerosol components that are primarily responsible for the absorption of solar radiation. (Note that we use the term “black carbon” to represent all the

solar radiation absorbing carbonaceous particles. As pointed out by a number of authors, the term is somewhat ambiguous since it does not differentiate between graphitic-like carbon particles (sometimes called EC for elemental carbon) and amorphous carbon particles. Perhaps a better term is LAC for light absorbing carbonaceous particles, however, we use black carbon in this paper for simplicity.)

2. Radiation Measurements and Modeling

[5] During the ACE-Asia intensive field study the NASA Solar Spectral Flux Radiometer (SSFR) was deployed on the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter to measure upwelling and downwelling solar spectral irradiance. The SSFR-measured irradiance has an absolute uncertainty of 1–3% over the wavelength range from 300 to 1700 nm. There is additional error associated with variation in the aircraft pitch and roll. For a recent description of the instrument, the calibration and a discussion of errors, see Pilewskie *et al.* [2003].

[6] The NASA Ames Airborne Tracking 14-channel Sunphotometer (AATS-14) was aboard the Twin Otter to measure the transmission of the direct solar beam in 14 channels (354 to 1558 nm). The data obtained by the AATS-14 during the ACE-Asia study period are presented by Schmid *et al.* [2003]. The AATS-14 is an enhanced version of the AATS-6 instrument, which flew on the C-130 aircraft during ACE-Asia [Redemann *et al.*, 2003]. The methods for data reduction, calibration, and error analysis have been described previously [see Schmid *et al.*, 2003].

[7] We developed a numerical radiative transfer model specifically for analysis of the SSFR data [Bergstrom *et al.*, 2003]. The major features of the model are the following: (1) k-distribution representation for O₂, O₃, CO₂ and H₂O absorption coefficients [Mlawer *et al.*, 1997]; (2) DISORT, a multiple scattering code [Stamnes *et al.*, 1988]; (3) Kurucz representation of the solar spectrum [Kurucz, 1992]; (4) filter functions from the SSFR [Pilewskie *et al.*, 2003].

[8] The model contains 140 bands of 10 nm width from 300 to 1700 nm matching the spectral coverage of the SSFR. The inputs are the vertical profiles of the gases, aerosol and clouds; as well as the spectral scattering and absorption properties of the aerosols and clouds. The spectral surface reflectance and the solar angle are also inputs. We simplify the aerosol scattering by assuming a Henyey-Greenstein phase function. This required specifying only the first moment of the phase function, the asymmetry parameter g . Further details about the radiative transfer model are given by Bergstrom *et al.* [2003].

[9] To calculate the solar irradiance in an aerosol layer, the optical depth, the single-scattering albedo (ratio of scattering to extinction) and the scattering distribution function of the aerosols must be specified. In the cases presented here the aerosol optical depth was known from the AATS-14 measurements. For the scattering distribution function we simply specified the asymmetry parameter over the solar spectrum as the radiative transfer model results for aerosol absorption have been shown to be relatively insensitive to the details of the aerosol scattering distribution

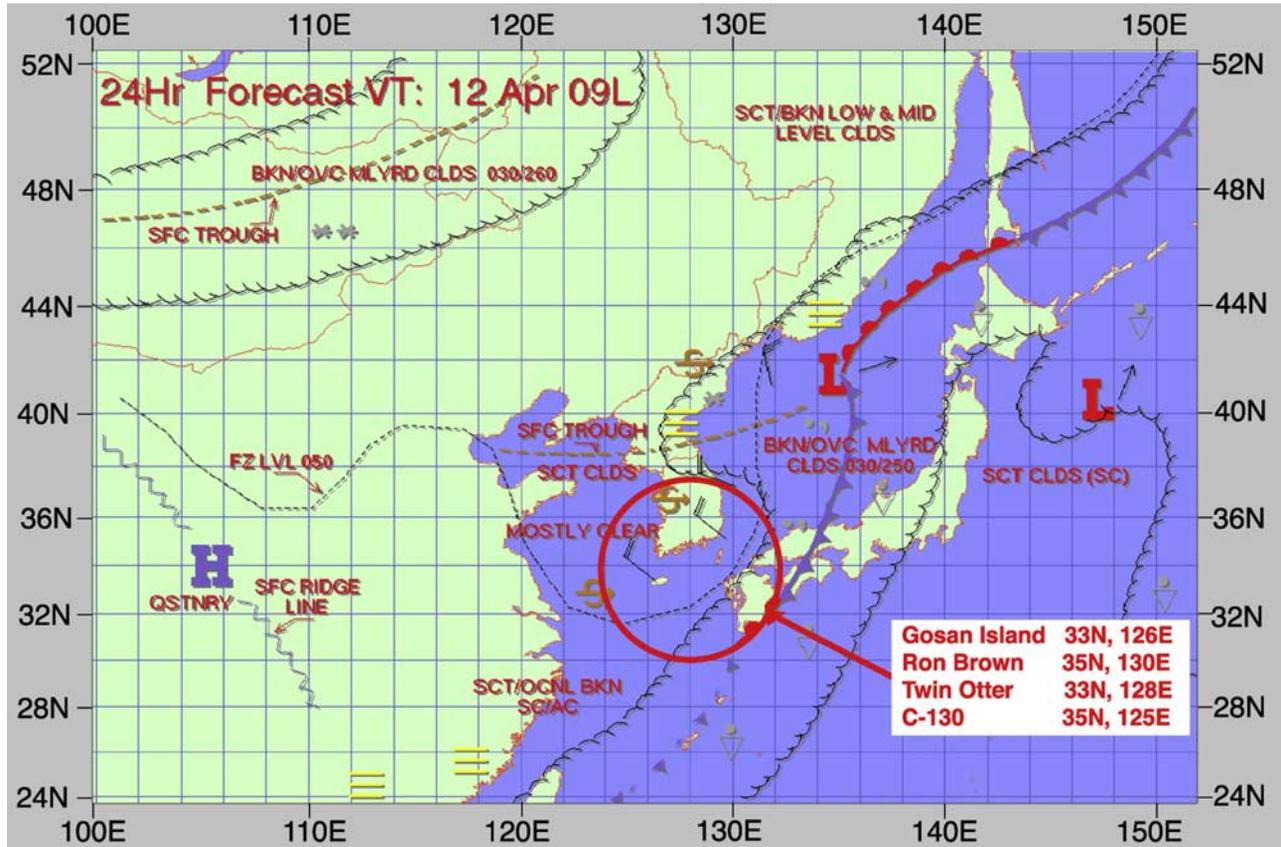


Figure 2. Location of the *Ron Brown*, the *Twin Otter*, and the *C-130* on 12 April 2001.

function [Bergstrom *et al.*, 2003]. Then the solar irradiance was calculated for a number of different values of the aerosol single-scattering albedo. In this way the aerosol effects on the solar radiation could be calculated without specifying a specific aerosol size distribution and refractive indices. By comparing the calculations at various aerosol single-scattering albedos with the measurements we are then able to estimate the aerosol absorption properties at each wavelength.

[10] A comparison of the measurements and model results for the solar irradiance at 2 km (above the aerosol layers) is shown in Figure 1 and indicates that the molecular scattering and absorption are reproduced by the model with high accuracy. This result was also obtained in the SAFARI study [Bergstrom *et al.*, 2003; Pilewskie *et al.*, 2003]. The average error between the measured and model solar irradiance was on the order of a few percent except in the regions of strong gas absorption.

[11] By measuring the upward and downward solar irradiance (when differenced yield the net solar flux) above and below an aerosol layer, we can determine the fractional absorption (α) of the layer [Pilewskie *et al.*, 2003]:

$$\alpha = \frac{(F_{\downarrow} - F_{\uparrow})_2 - (F_{\downarrow} - F_{\uparrow})_1}{F_{12}}$$

where F_{\downarrow} and F_{\uparrow} are the downward and upward irradiance, respectively, at the lower (subscript 1) and the upper (subscript 2) heights.

[12] The relationship of the single-scattering albedo, ω (the ratio of scattering to extinction), to the fractional absorption can be written formally as [Chandrasekhar, 1960, p. 11, equation (56)]

$$\alpha = \int_{\Delta\tau} (1 - \omega) \left[\int_{4\pi} I(\tau, \Omega) F_{12} d\Omega \right] d\tau \quad (1)$$

where I is the intensity (or radiance), Ω is the solid angle, and τ is the optical thickness at a particular wavelength. The single-scattering albedo is an important radiative property for aerosol climate effects since a small single-scattering albedo will lead to a warming effect, while a large single-scattering albedo will lead to a cooling effect.

[13] Equation (1) indicates that the fractional absorption is proportional to the co-albedo ($1 - \omega$). Since the spectral intensity is integrated over all angles, the fractional absorption is only weakly dependent on the scattering distribution function (or asymmetry factor). Therefore a large fractional absorption indicates a low single-scattering albedo while a small fractional absorption indicates a large single-scattering albedo (for nonabsorbing aerosols the single-scattering albedo is 1).

[14] As shown by Bergstrom *et al.* [2003] we can express the error in the single-scattering albedo in the limit of single scattering as

$$\delta\omega = (1 - \omega) \frac{\delta\alpha}{\alpha} + \frac{(1/\mu_0 e^{-\tau/\mu_0})}{(1 - e^{-\tau/\mu_0})} \delta\tau \quad (2)$$

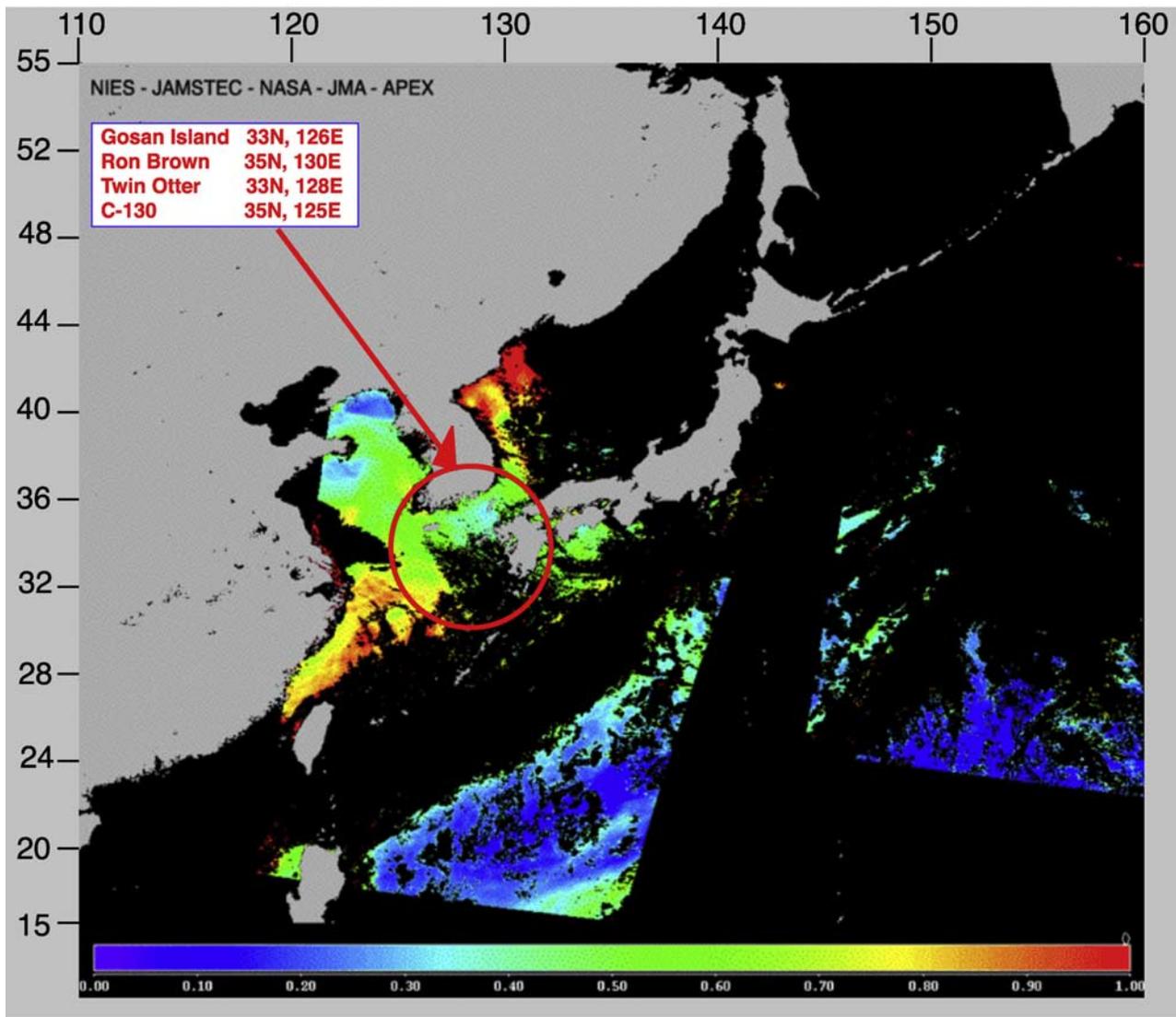


Figure 3. Optical depth map from the SeaWiFS analysis for 12 April 2001.

From equation (2) we can estimate the uncertainty in the single-scattering albedo from the uncertainty in the fractional absorption, α and the uncertainty in the optical depth, τ .

3. Moderately Thick Aerosol Layer: 12 April 2001

[15] On the morning of 12 April the Twin Otter flew from Iwakuni Marine Corps Air Station (MCAS) on the Island of Honshu near Hiroshima out into the Korean Strait to a location 33°N by 128°E, some 160 km east of the island of Gosan. The NOAA ship *Ronald H. Brown* was at 35°N by 130°E, about 400 km to the northeast of Gosan. The C-130 flew from Iwakuni MCAS into the Yellow Sea making several spirals near Gosan and continuing to the west coast of Korea then north over the Yellow Sea.

[16] The locations of the aircraft and the *Ron Brown* for 12 April are shown in Figure 2. (Figure 2 is from the meteorological forecast map issued before the flights on 12 April by J. Merrill of the University of Rhode Island.) The Twin Otter carried a number of aerosol sampling

instruments [Huebert *et al.*, 2003]. Mader *et al.* [2002] present elemental carbon, total organic carbon and carbonate mass from the aerosol sampling made aboard the Twin Otter. They measured an average value of elemental carbon of $0.56 \mu\text{g}/\text{m}^3$ for the 12 April flight. They also show that there appears to have been a significant amount of dust present as suggested by the amount of carbonate.

[17] Quinn *et al.* [2004] present optical depth and absorption results from measurements made aboard the *Ron Brown*. For 12 April they show aerosol optical depths of about 0.5 at 500 nm. Quinn *et al.* [2004] report that dust made up $70 \pm 20\%$ of the sub-10-micron aerosol mass during this period and EC made up less than 1%. The AERONET network (<http://aeronet.gsfc.nasa.gov/>) and Bush and Valero [2003] report aerosol optical depths at Gosan of about 0.3 at 500 nm as does Redemann *et al.* [2003] from AATS-6 aboard the C-130.

3.1. Atmospheric Conditions on 12 April 2001

[18] The ACE-Asia intensive period was marked by transport of pollution from east Asia and several mineral

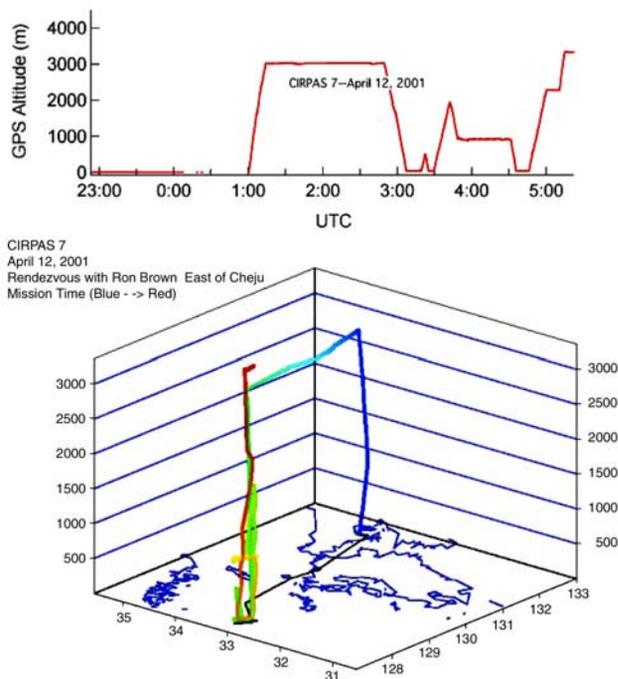


Figure 4. Twin Otter location and altitude for 12 April 2001.

dust outbreaks. The largest dust outbreak occurred on about 6 April in the inland Chinese deserts. Portions of the dust plume moved across the Pacific north of the Japanese Islands and reached the United States on 11 April [Jaffe *et al.*, 2003; Thulasiraman *et al.*, 2002]. Smaller amounts of the dust were transported south into the ACE-Asia region during the period of 10–13 April. The dust was widespread for several days including 12 April [Matsumoto *et al.*, 2003]. Several modeling studies [Chin *et al.*, 2003; Uno *et al.*, 2003] and also satellite images indicate that the dust traveled from the interior of China through the industrial regions of northeast China and Korea before it entered the Korean Strait. These models also indicate that the biomass burning in Southeast Asia did not contribute significantly to the black carbon observed in the Korean Strait during mid-April 2001 [Uno *et al.*, 2003].

[19] The meteorology of 12 April (see Figure 2) shows a low-pressure system pulling the aerosol through the Korean Strait region from west to east. The back trajectory analysis indicates that the lower boundary layer air in the Korean Strait came from central Asia and over Korea. The air apparently contained dust particles from the 6–7 April storm and then mixed with industrial pollution aerosol before it reached the Korean Strait.

[20] An aerosol optical depth analysis using the procedure of Higurashi and Nakajima [2002] for 12 April is shown in Figure 3. The SeaWiFS analysis yields an aerosol optical depth where the Twin Otter was located of about 0.35 at 500 nm. The analysis yields larger aerosol optical depth to the west of the Twin Otter, in particular at Gosan Island where the C-130 flew spirals and to the east where the *Ron Brown* was located on that day.

[21] The Twin Otter made several passes at various altitudes over the location 33°N, 128°E before returning

to Iwakuni, MCAS. The flight location and heights for 12 April 2001 are shown in Figure 4. Profiles of the multiwavelength aerosol optical depth and derived extinction coefficients from the AATS-14 aboard the Twin Otter are shown in Figure 5. The analysis performed by Schmid *et al.* [2003] using the AATS-14 extinction spectra indicated three layers: two lower layers (0–1.8 km) and an upper layer above about 2 km. The size of the particles can be estimated by the Angstrom exponent (the Angstrom exponent is the negative of the derivative of $\ln(\text{extinction})$ with respect to $\ln(\text{wavelength})$). For large particles (such as dust or sea salt) the extinction coefficient changes very little with wavelength so that the Angstrom exponent is small. On the other hand, the extinction coefficient of small particles (such as fresh combustion aerosols) falls off steeply with wavelength so that the Angstrom exponent is large [Bohren and Huffman, 1983]. Schmid *et al.* [2003] show that while the upper layer had small particles with a large Angstrom exponent, the total column optical depth was dominated by two lower layers with larger particles. The Angstrom exponent for the optical depth of the entire column was about 0.54 for wavelengths between 450 to 700 nm.

3.2. Measurements of the Spectral Solar Fluxes and Absorption

[22] The measured fractional atmospheric absorption (equation (1) above) for the aerosol layer between 43 m and 2700 m is shown in Figure 6, along with modeled spectra using two different values of the aerosol single-scattering albedo. The measured fractional absorption in the layer decreases from greater than 0.1 at 350 nm to 0.04 in the midvisible and stays relatively low in the region between the gas absorption bands in the near infrared. Comparison of the measured absorption spectrum with the modeled spectra shows that the single-scattering albedo must be ~ 0.8 in the near UV and increase with wavelength. That is, the modeled absorption for a single-scattering albedo of 0.8 agrees with the measurements at 350 nm and the modeled absorption for a single-scattering albedo of 0.99 is fairly close to the measurements at wavelengths longer than about 650 nm.

[23] The most prominent features in the absorption spectrum are the gas absorption bands. The water vapor bands centered at 942, 1135, and 1380 nm cause large peaks in the spectra. Retrieving the aerosol absorption is most reliable in the regions away from the gas absorption bands. In particular the regions of 350–700 nm, 800–900 nm, 1000–1100 nm, 1200–1300 nm, and 1500–1600 nm are regions least influenced by gaseous absorption.

[24] The only two common aerosol components that absorb solar radiation are black carbon (or light absorbing carbon) and dust (due primarily to hematite). Other aerosol components, sulfate, nitrates, liquid water absorb very little solar radiation [Bohren and Huffman, 1983, p. 438, Figure 14.1]. Black carbon and dust usually have different wavelength dependencies of the single-scattering albedo. The single-scattering albedo for a black carbon-industrial pollution mixture usually decreases with wavelength while dust absorption tends to increase with wavelength [Bergstrom *et al.*, 2002, 2003; Dubovik *et al.*, 2002]. Since there were indications of both black carbon and dust in the area on 12 April, an important question is whether or not we

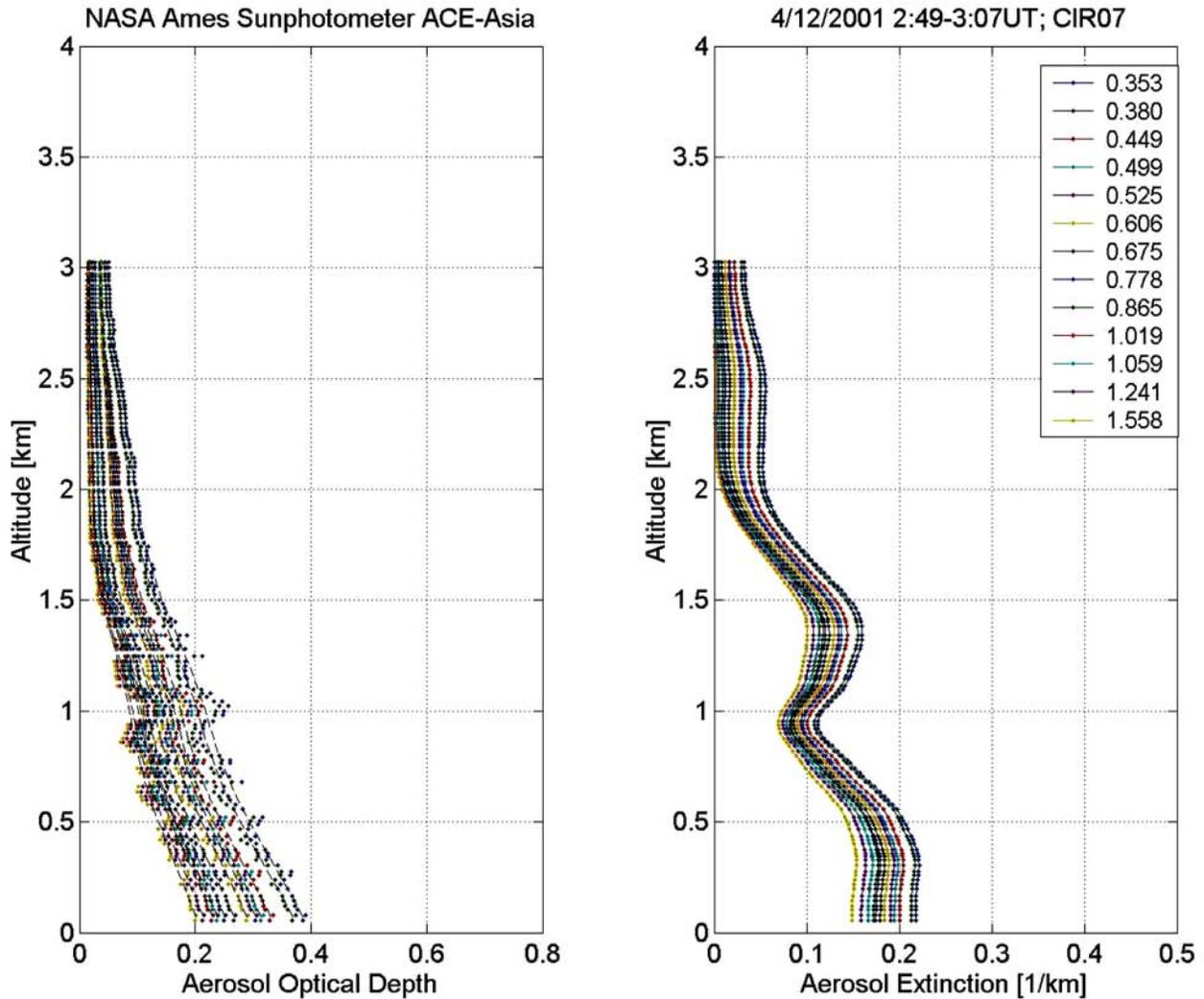


Figure 5. Vertical profiles of aerosol optical depth and extinction coefficient from the AATS-14 aboard the Twin Otter for 12 April 2001.

can estimate the relative contributions of each one from the wavelength dependence of the single-scattering albedo.

3.3. Estimates of the Spectral Single-Scattering Albedo and Absorption Optical Depth

[25] The single-scattering albedo $\omega(\lambda)$ for the layer can be derived from the measured fractional absorption $\alpha(\lambda)$ and the optical depth $\tau(\alpha)$ by interpolating the model results at each λ to make the model $\alpha(\lambda)$ equal the measured $\alpha(\lambda)$. The single-scattering albedo result for 12 April is shown in Figure 7. The single-scattering result from the four channel SeaWiFS analysis of *Higurashi and Nakajima* [2002] for 12 April at 550 nm at the Twin Otter location is also plotted.

[26] The derived single-scattering albedo has a minimum in the UV at 0.80 and increases through the visible to a relatively constant value of 0.95. Although there was considerable black carbon in the atmosphere sampled on this flight [*Mader et al.*, 2002] the wavelength dependence of the single-scattering albedo does not have a typical black carbon wavelength dependence [*Bergstrom et al.*, 2002,

2003; *Dubovik et al.*, 2002]. The wavelength dependence in Figure 7 is similar to a mineral dust single-scattering albedo [*Dubovik et al.*, 2002; *Sokolik and Toon*, 1999]. The values for Persian Gulf desert dust from *Dubovik et al.* [2002] are shown in Figure 7 for comparison. The *Dubovik et al.* [2002] values increase from 0.93 to 0.97 while our single-scattering albedo values increase from 0.87 to 0.95 in the same wavelength region. To better understand the spectral behavior of the single-scattering albedo, we plot the aerosol extinction optical depth and absorption optical depth for 12 April in Figure 8.

[27] The wavelength dependence of the single-scattering albedo depends on the wavelength dependence of both the extinction and the absorption ($\omega = 1 - \tau_{\text{abs}}/\tau$). If the extinction optical depth and absorption optical depth are approximated by power laws, i.e.,

$$\tau \sim \lambda^{-a}; \quad \tau_{\text{abs}} \sim \lambda^{-b}, \quad \text{where } \tau \gg \tau_{\text{abs}}, \quad (3)$$

then for $a > b$ the single-scattering albedo will decrease with wavelength and for $b > a$ the single-scattering

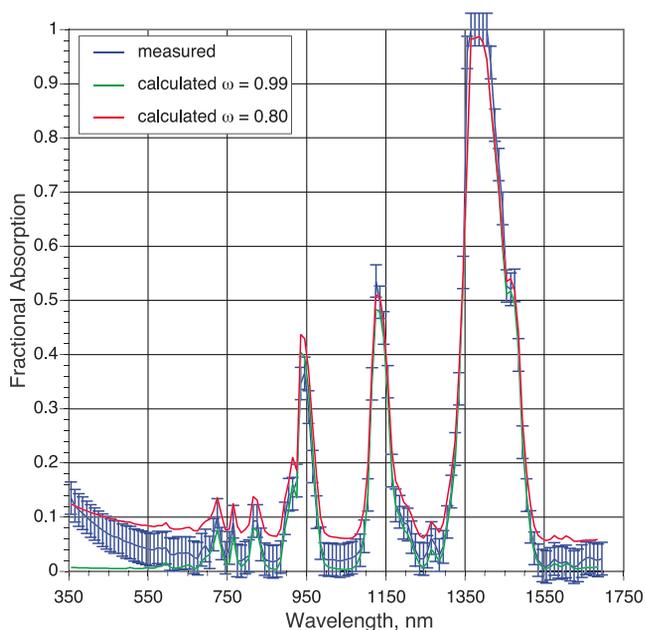


Figure 6. The 12 April measured and calculated fractional absorption for the layer between 43 m and 2700 m.

albedo will increase with wavelength. The curve of the derived single-scattering albedo in Figure 7 indicates that for the 12 April case $b > a$. This is shown in Figure 8 where a power law fit to the optical depth is approximately $\lambda^{-0.45}$ while the absorption optical depth falloff is $\lambda^{-2.0}$.

[28] As mentioned above, the Angstrom exponent (the quantity a in equation (3) above) indicates the relative size of the particles. A value of 0.45 for the fit to the optical

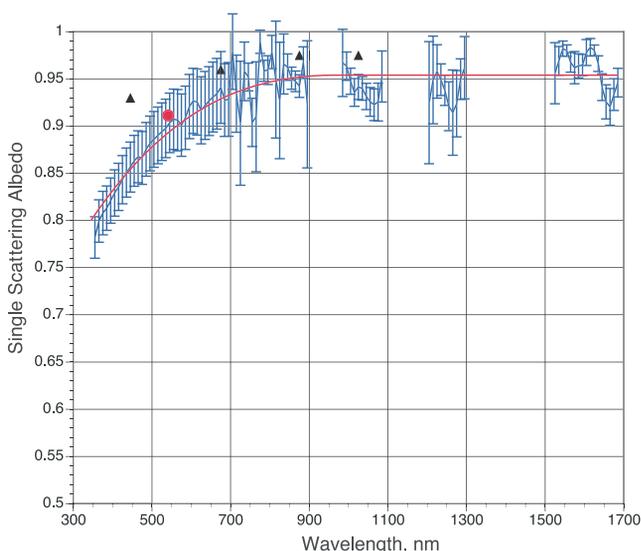


Figure 7. Derived aerosol single-scattering albedo. The smooth curve is a fit to the results. Black triangles, Persian Gulf dust [Dubovik *et al.*, 2002]. Red circle, SeaWiFS analysis for 12 April (this work).

depth indicates that the particles between 43 m and 2700 m altitude were fairly large, most likely containing dust particles as reported by Quinn *et al.* [2004]. Additionally, data from Gosan indicate that the dust transported from the inland deserts reached the surface around 0300 UT on 10 April and persisted until about 0000 UT on 14 April [Schmid *et al.*, 2003].

[29] It is difficult to tell from just the wavelength dependence of the absorption optical depth in Figure 8 whether the absorption was caused solely by black carbon particles or a combination of dust and black carbon particles. In general, most of the spectral absorption results in the literature for black carbon particles indicate a power law exponent of 1 to 2, where 1 is the small particle limit for wavelength-independent refractive indices. Bergstrom *et al.* [2002, 2003] found a value of 1 for pollution over the North Atlantic and a value of 1.3 for biomass burning particles in South Africa. However, Bond *et al.* [2002] show that the absorption from carbon particles produced from residential fuel use can have a power law exponent between 1 and 2.9. Schnaiter *et al.* [2003] found that while diesel soot particles have an absorption wavelength dependence of λ^{-1} , spark-gap-generated black carbon particles have a wavelength-dependent absorption of λ^{-2} . Thus a wavelength dependence of 2.0 could be caused by black carbon particles alone. (Recently, Kline *et al.* [2003] reported a wavelength dependence of λ^{-1} for the aerosol absorption from plumes from Asian population centers measured at Amami Ohshima, Japan.)

[30] For the measured black carbon particles to account for all the measured absorption, they would also have to have a relatively high mass absorption coefficient in the

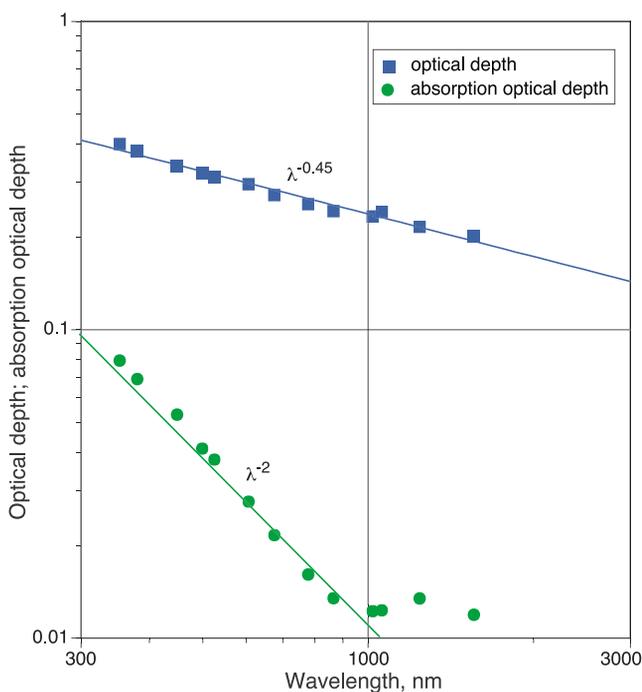


Figure 8. Wavelength dependence of aerosol optical depth and derived aerosol absorption optical depth.

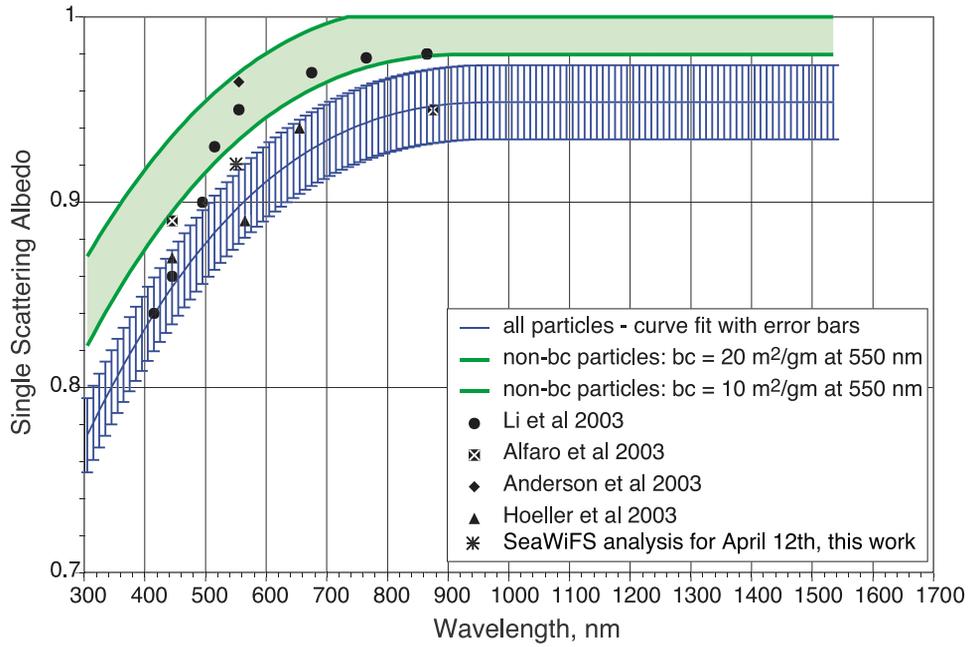


Figure 9. Estimated range of single-scattering albedo for mixture and for the nonblack carbon particles.

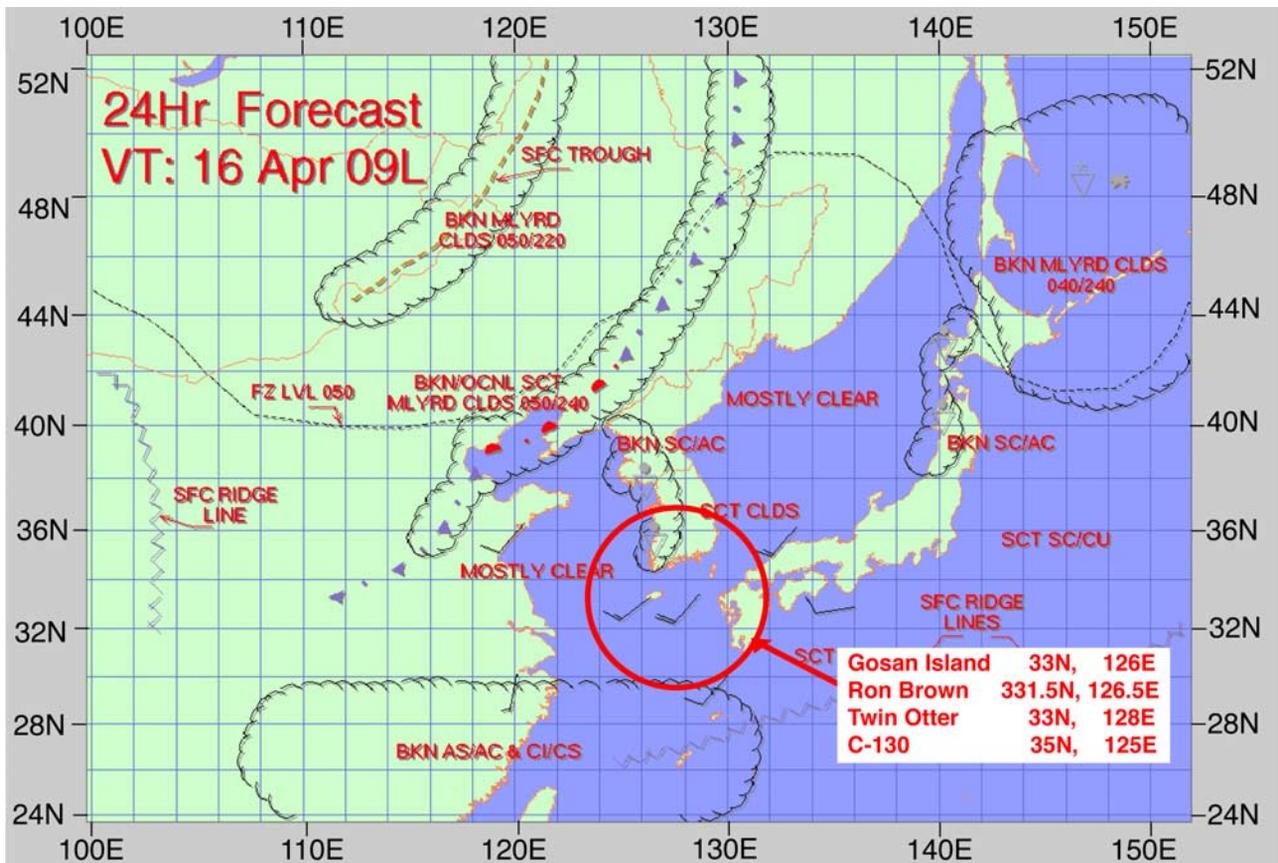


Figure 10. Location of the *Ron Brown*, the *Twin Otter*, and *C-130* for 16 April 2001.

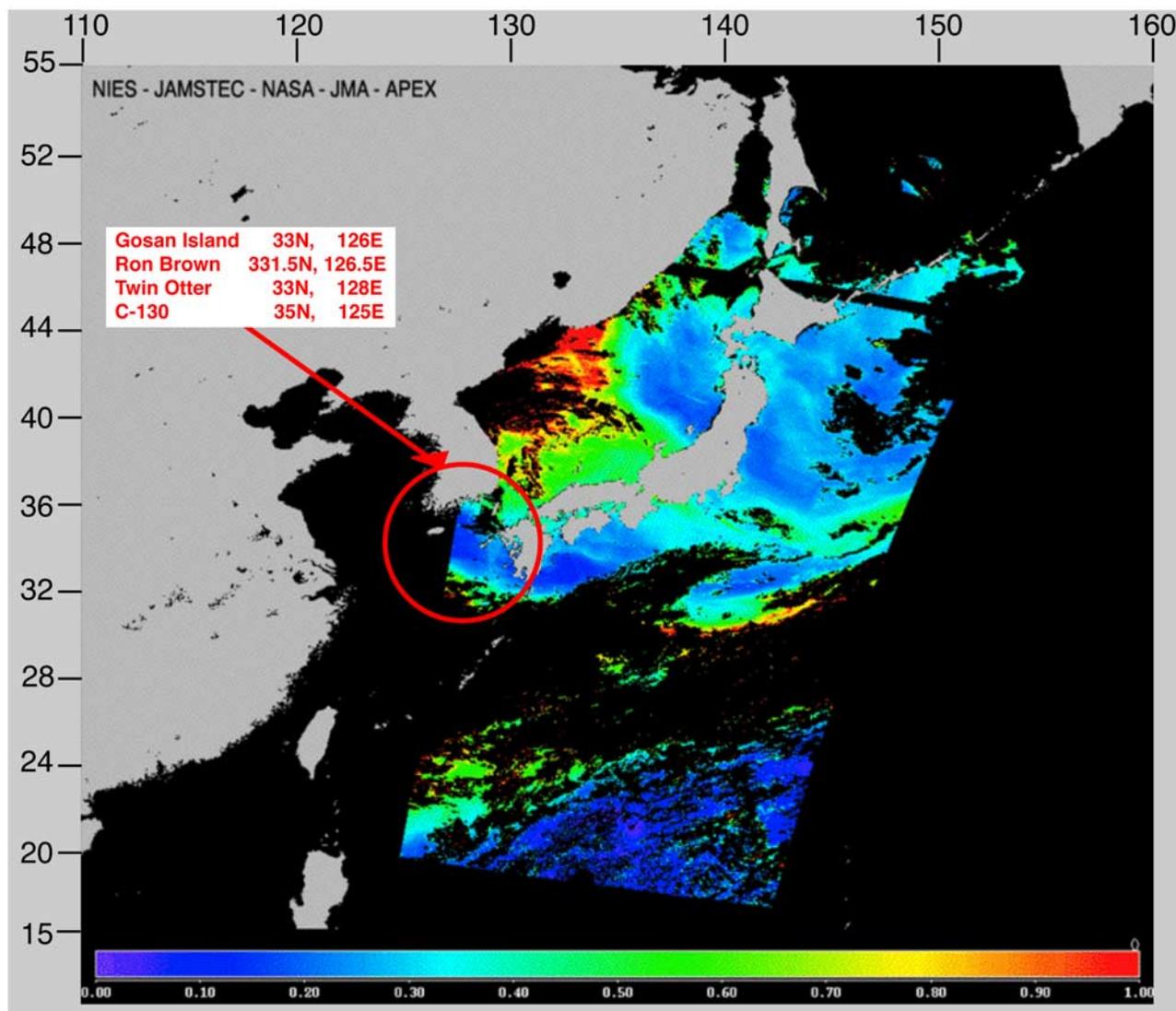


Figure 11. Optical depth map from the SeaWiFS analysis for 16 April 2001.

midvisible, approximately $25 \text{ m}^2/\text{g}$. A number of ACE-Asia investigators (*Chuang et al.* [2003] at Gosan, *Clarke et al.* [2004] on the C-130, *Anderson et al.* [2003] on the C-130, *Mader et al.* [2002] on the Twin Otter, and *Quinn et al.* [2004] on the Ron Brown) found a range of the mass absorption coefficient of black carbon particles to be between 5 and $25 \text{ m}^2/\text{g}$. However, values of the mass absorption coefficient above $15 \text{ m}^2/\text{g}$ are very difficult to explain theoretically. *Quinn et al.* [2004] showed that an internal mixing model could not account for the largest mass absorption coefficient values and *Chuang et al.* [2003] found that relatively complicated black carbon-dust geometries also did not affect the absorption properties significantly (similar to the results of *Fuller et al.* [1999] and *Reimer et al.* [2003]). This relatively large range of mass absorption coefficient results for black carbon is not currently understood. While it has been attributed to difficulties in measuring black carbon mass, to variations in the molecular structure of black carbon and to complicated particle geometry, the actual causes of

the variation remain uncertain. It is apparent, however, that different sources can produce black carbon particles with different mass absorption coefficients.

[31] The average values for the mass absorption coefficients from ACE-Asia are actually very similar between investigators. *Clarke et al.* [2004] and *Mader et al.* [2002] report a mean value for the ACE-Asia period for mass absorption coefficients of black carbon particles of about $10 \text{ m}^2/\text{g}$ at 550 nm. *Chuang et al.* [2003] have 12–15 m^2/g average value at Gosan, $\sim 10 \text{ m}^2/\text{g}$ for 12 April at 550 nm. *Quinn et al.* [2004] report the mass absorption coefficients for the period considered here was about $12 \text{ m}^2/\text{g}$.

[32] *Mader et al.* [2002] measured an aerosol absorption coefficient of 15 Mm^{-1} at 550 nm for 12 April and attributed it all to black carbon deriving a mass absorption coefficient of $27 \text{ m}^2/\text{g}$. Thus a power law exponent of 2.0 and a mass absorption coefficient of $27 \text{ m}^2/\text{g}$ would account for all of the aerosol absorption that we observe since our mass absorption coefficient was 14.7 Mm^{-1} at 550 nm. However, *Bond et al.* [2002] show that

a high mass absorption coefficient correlates with a low power law exponent value. They attribute this behavior to a greater degree of graphitization in the carbon particles that have a high mass absorption coefficient (such as diesel exhaust). Therefore it appears inconsistent to assume a high mass absorption coefficient and a large power law exponent.

[33] We can attempt to separate out the absorption due to black carbon and the other particles including dust by assuming a range of mass absorption coefficients and a wavelength dependence for the black carbon particles. By subtracting out the black carbon contribution from the total absorption we can estimate the absorption optical depth and single-scattering albedo for the remaining, nonblack carbon particles.

[34] The results of these calculations for the single-scattering albedo are shown in Figure 9. The range of black carbon mass absorption coefficient values used was 10 to 20 m²/g, and the wavelength dependence was assumed to be λ^{-1} . The result is that the single-scattering albedo for the nonblack carbon particles in Figure 9 is higher than that for the total mixture. This indicates that the remainder of the aerosol mixture is less absorbing than the black carbon particles. Also, the remainder absorbs primarily in the UV and relatively little in the visible and near-IR wavelength regions.

[35] The area between the upper curves in Figure 9 is shaded to indicate the likely range of possible single-scattering albedos for the remaining particles. We have also plotted several other single-scattering values for aerosols in the east Asia region for comparison. The single-scattering values from *Li et al.* [2003] are for Chinese soil particles, those from *Alfaro et al.* [2003] are from Sun photometer data at the ACE-Asia supersite in Zhenbeitai, China, for dust events, those from *Anderson et al.* [2003] are from the PSAP results for the dust cases observed aboard the C-130, and those from *Höller et al.* [2003] are for integrating sphere results for measurements at Yasaka, Japan (see below). The data point labeled SeaWiFS analysis is from the four-channel algorithm of *Higurashi and Nakajima* [2002] for 12 April at the Twin Otter location.

4. Thin Aerosol Layer Case: 16 April 2001

[36] The second case that we have examined occurred on 16 April 2001. The optical depth was one of the smallest during the ACE-Asia period [*Schmid et al.*, 2003]. On that day the Twin Otter again flew profiles under cloud-free conditions in the Korean Strait. The locations of the plane and ship are shown in Figure 10. The Twin Otter was in the same region as it was as on 12 April while the *Ron Brown* ship was farther to the south of Gosan.

4.1. Atmospheric Conditions for 16 April 2001

[37] By 16 April most of the dust had been removed from the atmosphere and the area became relatively clean. The SeaWiFS optical depth analysis for 16 April is shown in Figure 11. As shown the optical depth was estimated to be quite low (~ 0.1). The flight plan and vertical levels for the Twin Otter are shown in Figure 12. The measured optical depth from the AATS-14 (shown in Figure 13) is in general agreement with the SeaWiFS analysis. The extinction pro-

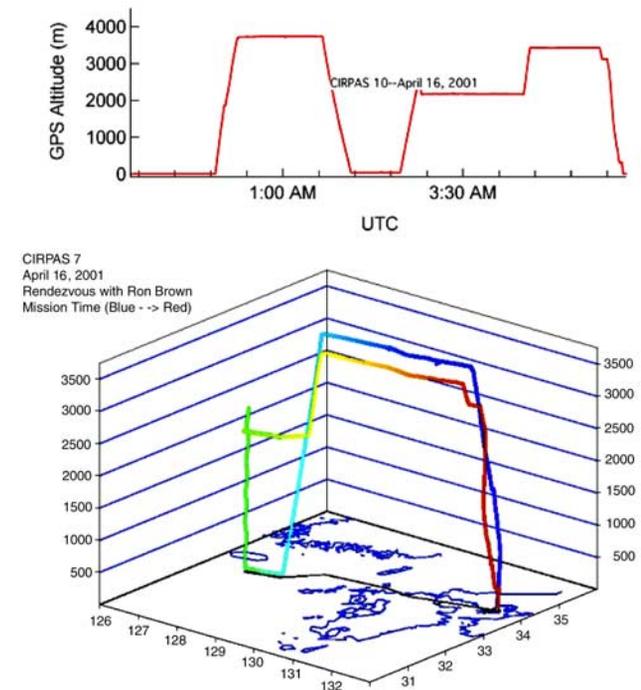


Figure 12. Flight path of the Twin Otter for 16 April 2001.

file shows a layer of aerosol between the surface and about 1 km.

4.2. Measurements of the Spectral Solar Fluxes and Absorption and Estimates of the Spectral Single-Scattering Albedo and Absorption Optical Depth

[38] The measured and two computed normalized absorption spectra are shown in Figure 14. The fractional absorption is quite low throughout the visible spectrum. The measured value is only slightly above modeled values using a single-scattering albedo of 0.99. The estimated single-scattering albedo of the aerosol is shown in Figure 15. The single-scattering albedo shows a decrease in the visible (with large error bars). The single-scattering result from the four channel SeaWiFS analysis of *Higurashi and Nakajima* [2002] for 16 April at the Twin Otter location is also plotted. The agreement with the single-scattering value from the SeaWiFS analysis is actually quite good as it was for the 12 April case.

[39] While the uncertainty in the measured absorption is larger than the absolute value, the derived spectral variation of the single-scattering albedo (decreasing with wavelength) shown in Figure 15 is suggestive of absorption by black carbon particles [*Bergstrom et al.*, 2002, 2003; *Dubovik et al.*, 2002; *Eck et al.*, 2003; *Höller et al.*, 2003]. Unfortunately, no measurements of black carbon or dust were made on 16 April aboard the Twin Otter so we cannot attempt to estimate the relative amounts of absorption by black carbon and by the other particles. *Chuang et al.* [2003] report black carbon measurements at Gosan on 16 April of approximately 0.8 $\mu\text{g}/\text{m}^3$ and an absorption coefficient of about 6 Mm^{-1}

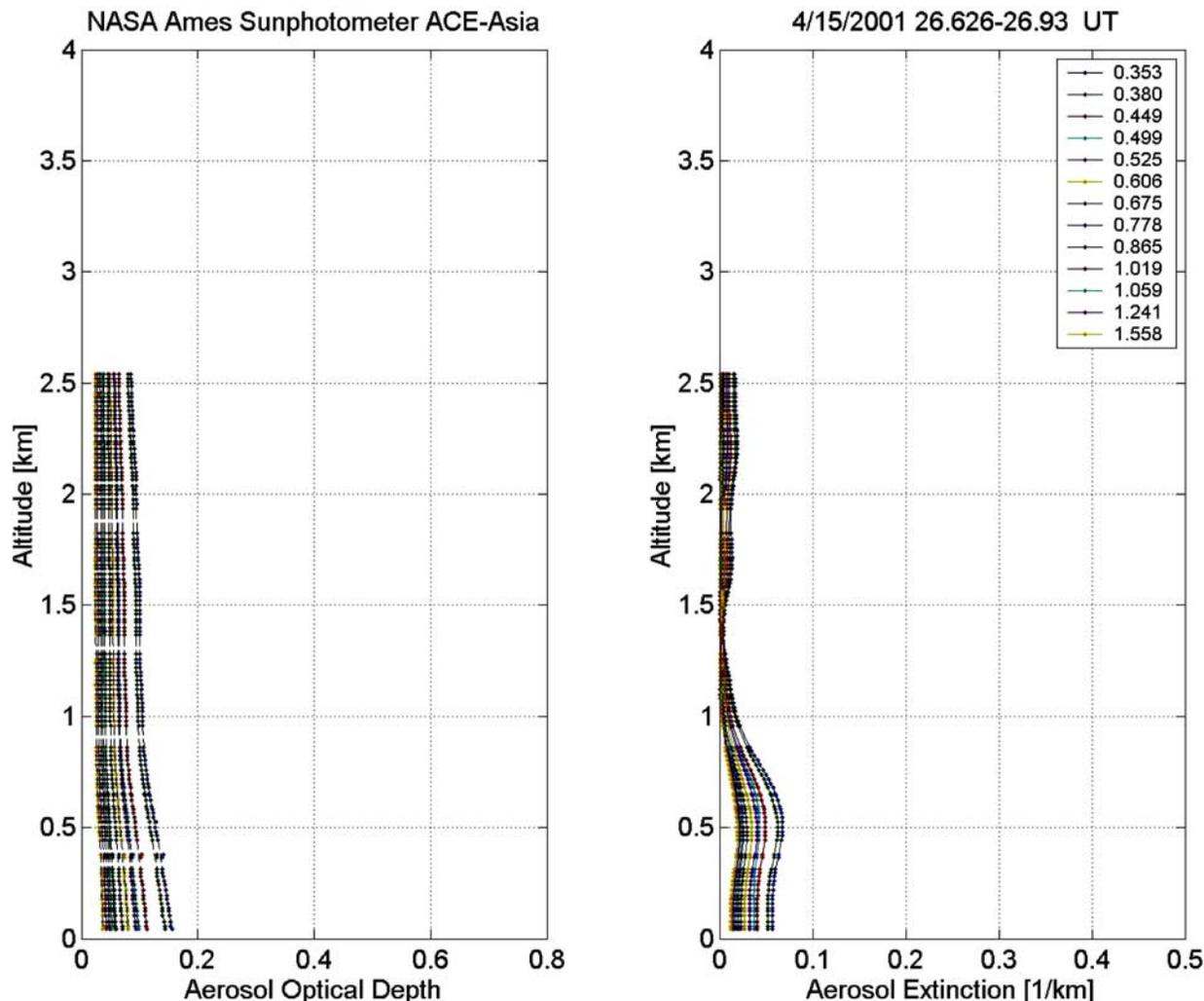


Figure 13. Aerosol optical depth and extinction coefficients from the AATS-14 aboard the Twin Otter for 16 April 2001.

at 550 nm. An EC concentration of $0.5 \mu\text{g m}^{-3}$ and an absorption coefficient of about 4 Mm^{-1} was measured on the *Ron Brown* [Bates *et al.*, 2004; Quinn *et al.*, 2004]. In addition, EC made up about 2% of the sub- $10\text{-}\mu\text{m}$ aerosol mass of the particles measured on the *Ron Brown*. These results are consistent with the absorption being caused by black carbon particles.

5. Comparison to Other Aerosol Absorption Measurements

[40] For the 12 April case our estimated total aerosol absorption coefficient at 550 nm is 14.7 Mm^{-1} that agrees well with Mader *et al.* [2002] who reported 15 Mm^{-1} . On the *Ron Brown*, Quinn *et al.* [2004] measured 12 Mm^{-1} . Chuang *et al.* [2003] report a relatively large value of 27 Mm^{-1} at Gosan, however, it is a 24 hour average. Quinn *et al.* [2004] report a single-scattering albedo of 0.94 ± 0.1 for the morning of 12 April. This value is based on measured scattering and absorption coefficients at 55% RH. Using simultaneously measured $f(\text{RH})$ for the scatter-

ing coefficient [Carrico *et al.*, 2003], the single-scattering albedo was adjusted to ambient RH. This compares to our derived single-scattering albedo of 0.90 ± 0.02 at 550 nm.

[41] For 16 April case we estimate an absorption coefficient of 5 Mm^{-1} . On the *Ron Brown*, Quinn *et al.* [2004] measured 4 Mm^{-1} while Chuang *et al.* [2003] measured about 6 Mm^{-1} . For such a small amount of aerosol and the large horizontal variability seen in ACE-Asia [Anderson *et al.*, 2003; Redemann *et al.*, 2003], the agreement is fairly good.

[42] As mentioned above, there were a large number of aerosol measurements made during the ACE-Asia intensive period. Most of the absorption measurements were made at one wavelength, but Höller *et al.* [2003] measured the aerosol single-scattering albedo at 450, 550, and 700 nm at Yasaka, Japan, during three weeks in March–April 2001. They found that during a dust episode (20–23 March 2001) the single-scattering albedo increased with wavelength while the period with the least amount of dust, the single-scattering albedo decreased with wavelength. Their average values at 550 were roughly 0.90 ± 0.05 for the dust period

and 0.92 ± 0.01 for the nondust period. The measurements were not explicitly corrected for relative humidity effects, but the samples were kept at a relative humidity below 35%. These values of single-scattering albedo are similar to the results found here.

[43] *Anderson et al.* [2003] made a large number of absorption and scattering measurements on the C-130 during the ACE-Asia period. They report mean values of 0.88 ± 0.03 for pollution and 0.96 ± 0.01 for dust single-scattering albedo. *Quinn et al.* [2004] observed an average value of 0.93 ± 0.03 at ambient relative humidity (determined as described above) and 550 nm. These values are also consistent with our findings.

6. Summary and Conclusions

[44] Analysis of the solar radiative flux measurements for 12 April 2001 resulted in a single-scattering albedo that was 0.8 at 400 nm and increased to 0.95 at about 900 nm and was relatively constant to 1700 nm. Our results and other ACE-Asia measurements suggest that the aerosol absorption was due to a mixture of black carbon and mineral dust. We attempted to estimate the amount black carbon absorption by assuming a range of values for the mass absorption coefficient. The absorption then attributed to the remaining particles shows significant UV absorption, but very little visible and near infrared absorption.

[45] The 16 April case was a low-optical-depth case with very little aerosol. The relative uncertainty is relatively large, but the aerosol appears to be very slightly absorbing. The spectral behavior of the absorption is consistent with black carbon particles from industrial pollution.

[46] Our results imply a relatively complicated aerosol mixture of both industrial pollution (including black carbon)

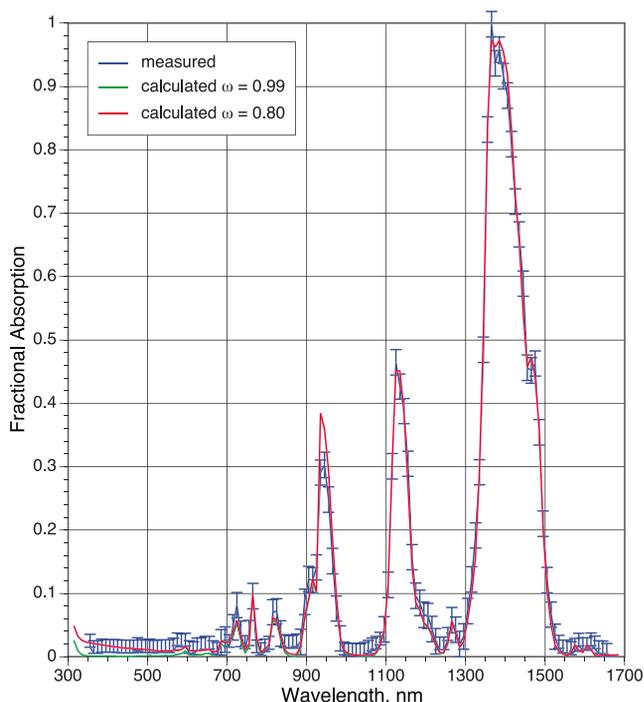


Figure 14. The 16 April measured and calculated fractional absorption.

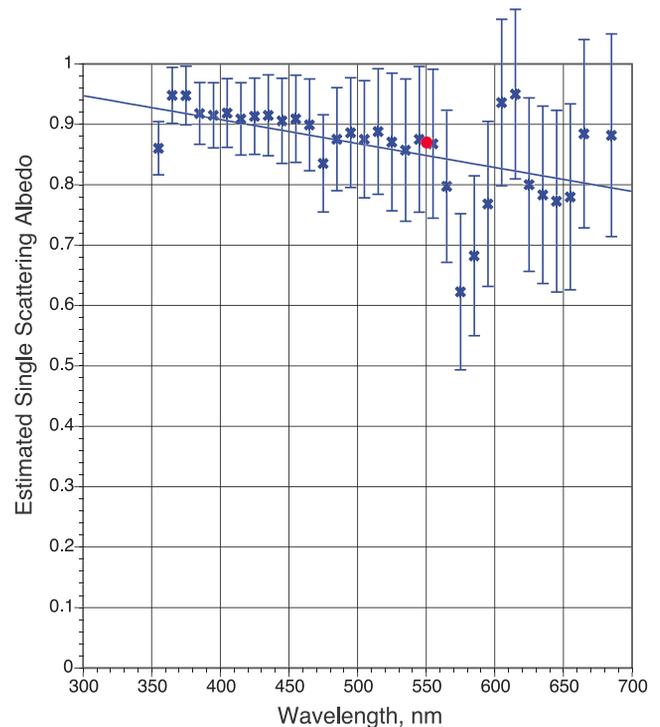


Figure 15. Estimated aerosol single-scattering albedo. Solid line is a fit to the results; red circle is SeaWiFS analysis for 16 April (this work).

and mineral dust in the ACE-Asia region. This underscores the need for careful measurements and analysis to separate out the absorption effects of mineral dust and black carbon in the east Asia region.

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